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Spin dynamics in the single molecule magnet Ni_4 under microwave irradiation[1]

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Quantum mechanical effects such as quantum tunneling of magnetization (QTM) and quantum phase interference have been intensively studied in single molecule magnets (SMMs). These materials have also been suggested as candidates for qubits and are promising for molecular spintronics. Understanding decoherence and energy relaxation mechanisms in SMMs is then both of fundamental interest and important for the use of SMMs in applications. Interestingly, the single-spin relaxation rate due to direct process of a SMM embedded in an elastic medium can be derived without any unknown coupling constant [1]. Moreover, nontrivial relaxation mechanisms are expected from collective effects in SMM single crystals, such as phonon superradiance or phonon bottleneck. In order to investigate the spin relaxation between the two lowest lying spin-states of the $S = 4$ single molecule magnet Ni_4, we have developed an integrated sensor that combines a microstrip resonator and micro-Hall effect magnetometer on a chip [2]. This sensor enables both real time studies of magnetization dynamics under pulse irradiation as well as simultaneous measurements of the absorbed power and magnetization changes under continuous microwave irradiation. The latter technique permits the study of small deviations from equilibrium under steady state conditions, i.e. small amplitude cw microwave irradiation. This has been used to determine the energy relaxation rate of a Ni_4 single crystal as a function of temperature at two frequencies, 10 and 27.8 GHz. A strong temperature dependence is observed below 1.5 K, which is not consistent with a direct spin-phonon relaxation process. The data instead suggest that the spin relaxation is dominated by a phonon bottleneck at low temperatures and occurs by an Orbach process involving excited spin-levels at higher temperatures [3]. Experimental results will be compared with detailed calculations of the relaxation rate using the density matrix equation with the relaxation terms in the universal form.

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